

New features in the stability and fission decay of superheavy Thorium isotopes

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Abstract:

Superheavy isotopes are highly neutron rich nuclei in the vicinity of neutron drip-line, stabilized by shell effect against the instability due to repulsive component of nuclear force, analogous to superheavy elements similarly stabilized against Coulomb instability. Here we discuss the stability and fission decay properties of such nuclei in the ^{254}Th region and show that they are stable against α and fission decay and have β -decay life time of several tens of seconds. In particular, the ^{254}Th nucleus has a low fission barrier and unusually large barrier width. This makes it an ideal thermally fissile nucleus, if formed by means of a thermal neutron, like other known nuclei such as ^{233}U , ^{235}U , ^{239}Pu in this actinide region. It shows a new mode of fast fission decay, which may be termed as multifragmentation fission, in which in addition to two heavy fragments large number of scission neutrons are simultaneously produced. Its likely synthesis during the r-process nucleosynthesis will have important bearing on stellar evolution, and here in the laboratory, it has great potential in energy production.

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The superheavy elements in $Z = 114 - 126$ region [1, 2, 3] which are vulnerable to spontaneous fission [4, 5, 6, 7, 8] due to the influence of repulsive Coulomb force of protons, have been predicted to be stable due to shell effect. Due to this effect, the periodic table has extended upto $Z=116$ [1] by now with promise to extend further along the valley of stability, thus elongating the stability peninsula [9, 10, 11]. Another effect that was shown by us earlier is the shell stabilisation of highly neutron rich superheavy isotopes of $Z = 62, 78$ and 90 nuclei [12]. It is known that the nucleon-nucleon force itself has a repulsive component (triplet-triplet, singlet-singlet) whose contribution progressively increases with the increase of neutron number making the nucleus unstable. The shell effect can stabilize this instability giving rise to new magic numbers in the vicinity of neutron drip-line. This is a complementary parallel effect which should widen the stability peninsula. In fact, in an extensive study involving three different methods, infinite nuclear matter (INM) model [13, 14, 15, 16], relativistic mean field (RMF) theory [17, 18, 19, 20, 21] and Strutinsky shell correction calculation [22], it was shown [12] that islands of stability around new magic numbers $N=164, Z=90$; $N=150, Z=78$; and $N=100, Z=62$ will exist giving rise to superheavy isotopes ^{254}Th , ^{228}Pt and ^{162}Sm . The N/Z ratios of these nuclei are 1.82, 1.92 and 1.62 respectively, in contrast to the value of 1.54 for the doubly closed shell superheavy element $^{298}\text{X}_{114}$ which is quite similar to that of ^{208}Pb and ^{235}U in the valley of stability. This result is depicted here in Figure 1. It may be recalled that a new frontier in 1980's has opened up with the production of radioactive ion beams with the prospect of synthesis of about 5000 nuclei in the laboratory, and thereby, extending the nuclear peninsula upto drip-line region. The ultra neutron rich nuclei are expected to show totally new properties and will probably be of immense utility to mankind. Here we investigate the properties of a representative ultra-neutron-rich nucleus ^{254}Th which is a superheavy isotope of thorium. It is known that ^{232}Th is a fertile material whose fission decay properties are well known. We show that ^{254}Th is stable against α -decay and fission decay and has β^- -decay half-life of tens of seconds. Most interestingly, ^{254}Th has unusual fission properties with low fission barrier of 3.57 MeV, but a very large barrier-width, which makes it infinitely stable against spontaneous

fission but fissionable if formed by means of a thermal neutron. It will undergo a new mode of fission decay in which, in addition to two fragments, large number of surplus neutrons will be instantaneously produced. This may be termed as multi-fragmentation fission. Due to highly stable character, this ^{254}Th nucleus will have important implications for r -process nuclear synthesis in stellar evolution and also great potential for energy production in the laboratory.

Alpha and Beta decay of Th isotopes

The usual magic nuclei with nucleon numbers 2, 8, 20, 50, 82 and 126 have their special properties, in particular extra stability, due to the close shell, and they occur along the central line in the valley of stability. However, as one goes across the valley towards the neutron drip line, as is shown in Figure 1, new shell closures can arise giving rise to new magic nuclei, and new islands of stability around them. The search and discovery of such nuclei is on the frontier of nuclear physics research today. It will be interesting to study the decay properties of the ultra neutron rich heavy nuclei. It is well known that, in general, heavy nuclei with $A > 150$ are unstable against α -decay while for lighter nuclei this process is improbable. Infact the superheavy nuclei $^{289}\text{X}_{114}$, $^{292}\text{X}_{114}$ show such decay, which seems to be the case for all heavy nuclei along the line of stability. The heavy neutron rich nuclei are expected to show deviation from this feature. In Figure 2a, the α -decay half-lives of different Th isotopes calculated using the predictions of the masses in INM [24] and Howard-Möller (HM)[25] mass models and RMF theory [26] are shown. The calculation of α -decay half-life T_α has been done using the widely used standard formula $\log T_\alpha / \text{sec.} = (aZ + b)Q_\alpha^{-1/2} + (cZ + d)$, where $a = 1.66175$, $b = -8.5166$, $c = -0.20228$ and $d = -33.9069$, due to Viola and Seaborg [27]. The value of Q_α is estimated by using the standard relation $Q_\alpha(Z, N) = BE(Z-2, N-2) + BE(2, 2) - BE(Z, N)$, where BE is the binding energy. In general, all the isotopes are quite stable against α -decay with decay life-time exceeding 10^{20} years for all the three mass models. The RMF and INM show a prominent peak around

$N = 160 \sim 168$ as mark of extra stability for isotopes around $N=164$. Beyond that the Q_α values become negative prohibiting such decay altogether. Thus ^{254}Th can be considered to be stable against α -decay.

The neutron-rich nuclei are generally β^- active. They will successively emit β^- particles until they reach the line of β -stability in the valley, a parallel scenerio to the superheavy elements which attend stability by successive α -emission (see Figure 1). The β -decay half-lives T_β are estimated by using the standard formula due to Seeger, Fowler and Clayton [28] $T_\beta = \frac{18 \times 10^5 \Delta \ln 2}{W_0^6}$ sec., where Δ = number of states of the daughter nucleus within 1 MeV of ground state nucleus $\times \exp(-A/290)$ with A being its mass number and $W_0 = BE(Z+1, A) - BE(Z, A) + 1.29$ MeV. The half-lives of Th isotopes so obtained are presented in Figure 2b, which stop falling rapidly from $N=164$ with some flat peaks thereafter indicative of extra stability, and the values are in the realm of tens of seconds. Thus ^{254}Th can be considered to be a fairly stable nucleus for laboratory studies.

Fission decay of ^{254}Th

The study of the fission decay properties of ^{254}Th is of special importance because the lighter stable isotope ^{232}Th is known to be a fertile nucleus. The question how the ultra-neutron-rich superheavy isotopes for the actinide nuclei will decay by fission has not been addressed before. It is here one may expect some exotic new phenomena. The fission barrier determines the fission decay properties of the nucleus. The nucleus can overcome the barrier and undergo decay when excited by an external agency like an energetic neutron incident upon it. The neutron separation energy S_n provides a measure of excitation for the fission decay process. We have plotted in Figure 3 the neutron separation energy S_n and fission barrier B_f of the even Th isotopes as a function of neutron number. The black squares and the green stars are the S_n values of the HM and INM models. They are connected by lines to guide the eye. The closed red circles represent the values of fission barrier B_f taken from Howard and Möller [25]. It is well known that if $S_n < B_f$, then the nucleus can not undergo thermal neutron fission. The

fission threshold $E_{nm} = B_f - S_n$ has to be overcome by impinging with an energetic neutron, and thereby the nucleus will undergo fission. Hence, if $S_n > B_f$, then thermal neutron (with practically zero energy) can cause fission. The thermal fission becomes a very attractive possibility for energy generation. It is interesting to see in Figure 3 that, although normal Th (^{232}Th) is not thermally fissile, its isotopes around $N=164$ in the range $N=162-170$ are thermally fissile, which has important implication and consequences. We have as yet only a few generally known nuclei which are fissionable with thermal neutron, out of which only one i.e., ^{235}U is naturally occurring with an abundance of 0.71% in natural uranium, and the others like ^{233}U and ^{239}Pu can be artificially produced. Of course they have long half-lives of 1.6×10^5 and 2.1×10^4 years respectively, but ^{254}Th as will be shown *a posteriori* is permanently stable against spontaneous fission.

Fission barrier profile of ^{254}Th

We now consider the nature of the fission decay mode of ^{254}Th , which is primarily governed by the profile of the fission barrier. The height and width of the fission barrier which is supposed to be parabolic in nature have to be obtained. We have followed an empirical method to get the width of the barrier from the systematics of the known experimental fission half-lives, and extrapolate them to extremely neutron rich region of interest to us. The fission half-lives can be calculated in a simplistic manner as $\tau_{1/2} = \ln 2 / np$, where n is the number of barrier assault by the decaying fragment, related to the width $\hbar\omega$ by $n\hbar = \hbar\omega / 2\pi$. And p is the penetrability of the barrier given by $p = [1 + \exp(2\pi B_f / \hbar\omega)]^{-1}$. Taking the values of B_f from reference [25], and using the experimental $\tau_{1/2}$ [29], we get the values of $\hbar\omega$, the systematics of which so obtained are shown in the plot of $\hbar\omega$ versus B_f in Figure 4 for various actinide nuclei. It is indeed very revealing as well as interesting that the plot shows a linear behaviour with progressively increasing slope with the increase of proton number of the elements from 90 to 96. For the next element Cf with $Z=98$, the linear behavior gets fuzzy. However, the mean

follows the trend with a higher inclination. The trend is more conspicuously restored for Fm with $Z=100$. This deviation correlates well with the fission mass yield systematics, where considerable deviation from the standard well-defined two peaks occur for Fm isotopes. It may be mentioned that such systematics of B_f versus $\hbar\omega$ has been analysed for the first time here. The width $\hbar\omega$ for any isotope with calculated fission barrier may be obtained by extrapolation of the linear graph. For ^{254}Th with a fission barrier of 3.57 MeV, we obtained the value of $\hbar\omega$ close to zero. Now we can construct the parabolic barrier of base width Δr somewhat schematically, using the value of $\hbar\omega$ following the relation $1/\hbar\omega = d^2V/dr^2$, where V is the potential energy. The barrier so obtained is presented as the blue curve in Figure 5, which is an extremely flat and wide barrier. For comparison, we have presented in the same figure the fission barrier of ^{232}Th as red curve constructed with $\hbar\omega = 0.415$ MeV obtained from the experimental half-life [29], and $B_f = 7.40$ MeV from Howard and Möller [25]. The unusually large fission width derived above in case of ^{254}Th is quite understandable and in conformity with expectation. It is not necessarily true that the fission barrier falls with the increase of neutron number in an isotopic chain, it may even rise [30]. So the wide barrier obtained here is rather specific to ^{254}Th , which may be related to its shell closure structure.

Our experience from the fission studies of uranium, has shown that the excess neutrons mainly inhabit the neck region when the nucleus deforms on its journey to fission, which is related to the width of the barrier. Since in the case of ^{254}Th , 22 excess neutrons participate compared to that in ^{232}Th , a larger neck must be formed giving rise to a flat and wider barrier. This feature is also in tune with the fact that in the limiting case of no Coulomb force, the barrier will disappear reminiscent of infinite width and zero height. This interesting feature of the barrier of ^{254}Th with low height but large width points out a new feature where, the nucleus is infinitely stable against spontaneous fission. But with a slight deposition of energy with a thermal neutron, it will undergo fission. Thus the fission half-life of spontaneous fission is nearly infinite.

New mode of fission decay

We next examine the decay mode of fission of a nucleus, (A, Z) decaying to (A_1, Z_1) and (A_2, Z_2) , solely guided by the Q -value systematics of the process, where Q is defined as $Q^f(A, Z) = BE(A_1, Z_1) + BE(A_2, Z_2) - BE(A, Z)$. As is well known, the major driving force for the decay is the Q -value of the reaction. The probability of fragment mass yield in a given channel is directly related to the Q -value. In Figure 6, we have plotted the Q -values of the binary decay into two fragments A_1 and A_2 , as a function of the mass number of the A_1 fragment, for all the relevant elements with even values of Z , starting from 46 to 58. The complimentary fragment (Z_2, A_2) is thereby fixed. Since the yield falls rapidly with the decrease in Q -value for an element, we have only shown the distribution of Q -values lying above 90% of the highest values. For the sake of comparison, the Q -value distributions for both ^{232}Th and ^{254}Th are shown in Figure 6. The black and red curves pertain to ^{254}Th and blue and green to ^{232}Th calculated using the predicted masses in INM and RMF models respectively. Since ^{254}Th is an ultra-neutron-rich isotope, during its scission many neutrons will be left free as they can not be bound to either of the fragments which may be already on the drip-line. In the figure, vertical lines mark the drip-line of the respective elements. As expected, the drip-line falls nearly in the middle of the distribution in case of ^{254}Th , indicating that all the isotopes lying to the right of it will be unstable against instantaneous neutron emission from the fragments at scission. These drip-line neutrons will be simultaneously produced along with the newly formed fragments. In the usual fission process, neutrons are produced by the emission from the fragments after they are fully accelerated. But in the present case of ^{254}Th , a certain number of neutrons will be simultaneously produced, signalling a new mode of fission decay which may be termed as *multi-fragmentation fission*. An order of magnitude of these prompt multi-fragmentation neutrons can be estimated from the mass yield plot for ^{254}Th as shown in Figure 6, which turns out to be about 2 to 3 neutrons per fission. These are the additional neutrons apart from the normal multiplicity of neutrons emitted from the fragments. In case of neutron-induced fission of normal ^{232}Th and ^{235}U nuclei, the fission neutron multiplicities are of the order 2.3 to 2.5 [33, 34]. This number is, therefore, nearly doubled in case of ^{254}Th fission,

which will have important implications on the energetics of the fission process.

Although, the beta decay life-time of ^{254}Th is few tens of seconds it is much larger than the nucleon decay life time, which is of the order of 10^{-17} seconds and therefore it will have a great implication in the r -process nuclear synthesis and consequently stellar evolution. We may also conjecture that in the laboratory, such nuclei can be of immense potential as energy source.

Conclusions

In conclusion, we have studied the decay properties of Th isotopes, in particular ^{254}Th , a superheavy isotope of thorium which has been predicted earlier to be a doubly close shell nuclei with $N=164$, and $Z=90$ stabilised by shell effect against repulsive nuclear force instability. It is stable against α -decay and has β -half-life of several tens of seconds. The fission decay properties of such neutron-rich nuclei in particular in the actinide region have not been addressed before, which has important bearing on the RIB programmes in many laboratories in the world seeking the synthesis of about 5000 nuclei in the exploration upto the drip-lines. On the basis of the systematics of neutron separation energies and fission barriers, it has been shown to have a rare property of being thermally fissile putting it into the rank of other such generally known nuclei like ^{233}U , ^{235}U and ^{239}Pu . Its fission barrier profile determined empirically using the systematics of experimentally known so far in this actinide region fission half-lives and barrier of the actinide nuclei, shows it to be very flat and wide. Such feature is in conformity with the expected long neck due to the excess number of neutrons to the tune of 22 over that of the ^{232}Th , presumably supported by its shell closure property. Coupled to this, its small barrier of 3.57 MeV, makes it extremely stable against spontaneous fission, but highly vulnerable to thermal neutron fission, a quite unique property indeed. A new mode of fission decay named multifragmentation fission is predicted for this nucleus, where in addition to two heavy fragments, 2 to 3 scission neutrons will be simultaneously produced. This is in addition to the normal multiplicity of neutron emitted by the

fragments which are of the order of 2.3 to 2.5. Thus the doubling of the neutron emission per fission will have strong implications as for the r -process nucleosynthesis in the stellar evolution. It also presents an attractive possibility as a source of energy production in the laboratory.

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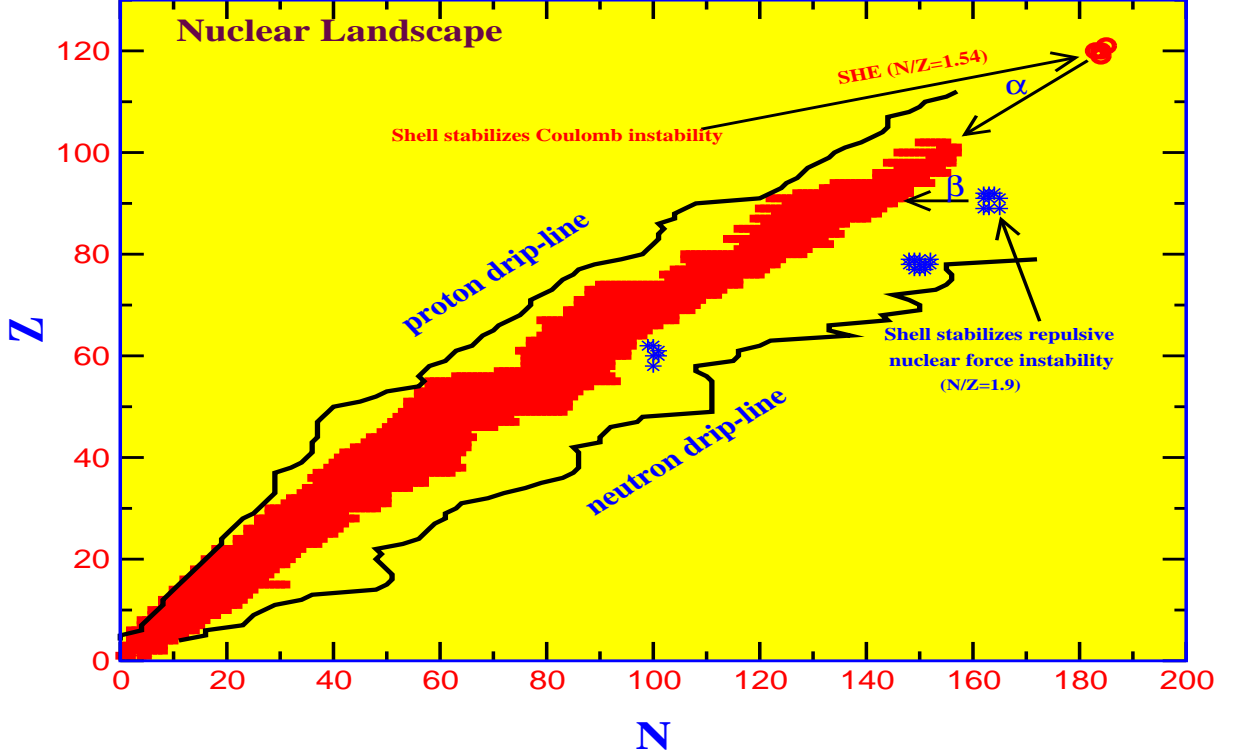


Figure 1: The new islands of stability identified around the magic numbers $N=100$, $Z=62$; $N=150$, $Z=78$ and $N=164$, $Z=90$ have been shown by the blue stars in the nuclear chart. The red area represents the nuclei with experimentally known binding energies [23]. The neutron and proton drip-lines shown as solid curves are the predictions of infinite nuclear matter model. The superheavy elements occur along the line of β -stability and is shown by red circles, whereas the superheavy isotopes occur close to the neutron drip-line. The superheavy elements undergo α -decay and the superheavy isotopes β -decay to attain higher stability. The shell effect stabilizes the superheavy elements against Coulomb instability while it stabilizes the superheavy isotopes against repulsive nuclear force instability.

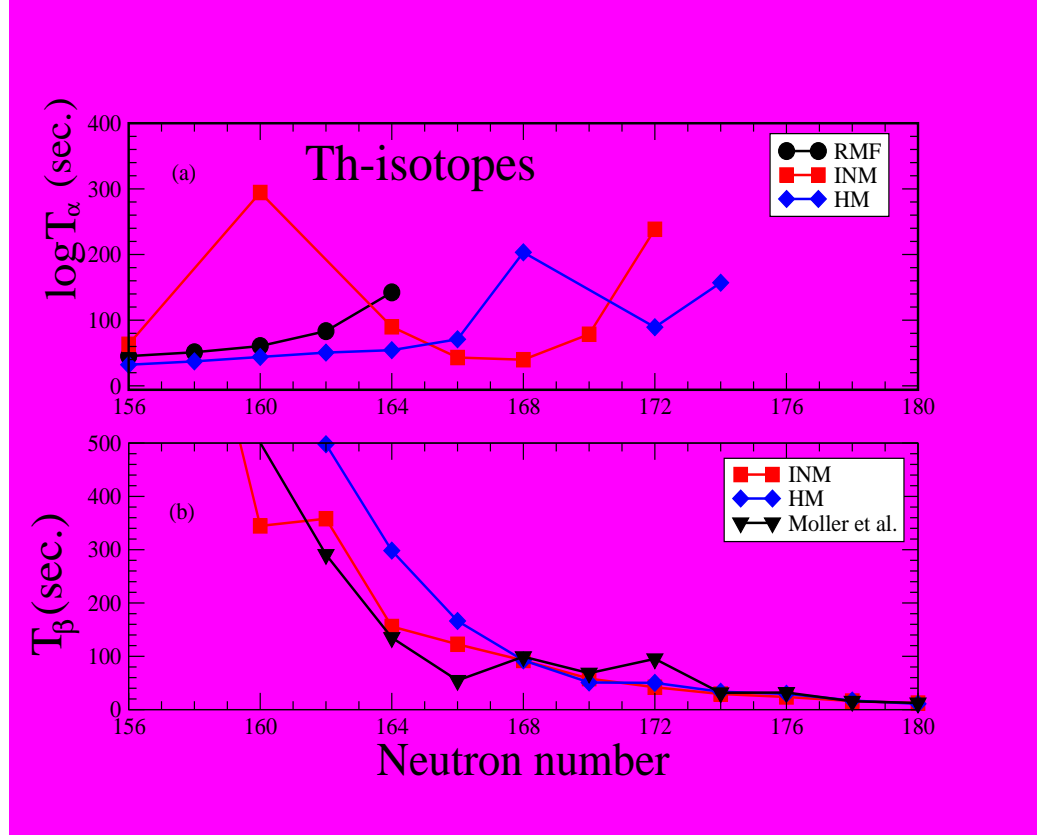


Figure 2: (a) α -decay half-life for Th isotopes using the empirical formula $\log T_\alpha = (aZ + b)Q_\alpha^{-1/2} + (cZ + d)$ of Viola and Seaborg [27]. The Q_α -value calculated by using the mass prediction of INM, RMF and HM models. The α -decay half-lives obtained by all the three models show similar trend. All the three models show a peak around $N = 160 - 170$ before Q_α becomes negative with increase of neutron number forbidding the process. From the y-axis it is clear that the Th isotopes are extremely stable against *alpha*-decay. (b) The β -decay half life for Th isotopes using the formula [31, 28] $T_\beta = \frac{18 \times 10^5 \Delta \ln 2}{W_0^6}$ sec. is calculated, where Δ = number of states of the daughter nucleus within 1 MeV of ground state nucleus $\times \exp(-A/290)$ with A being its mass number and $W_0 = BE(Z + 1, A) - BE(Z, A) + 1.29$ MeV. The value of the β -decay half-lives decreases almost exponentially with increase of neutron number. ^{254}Th has an appreciable life-time of tens of seconds.

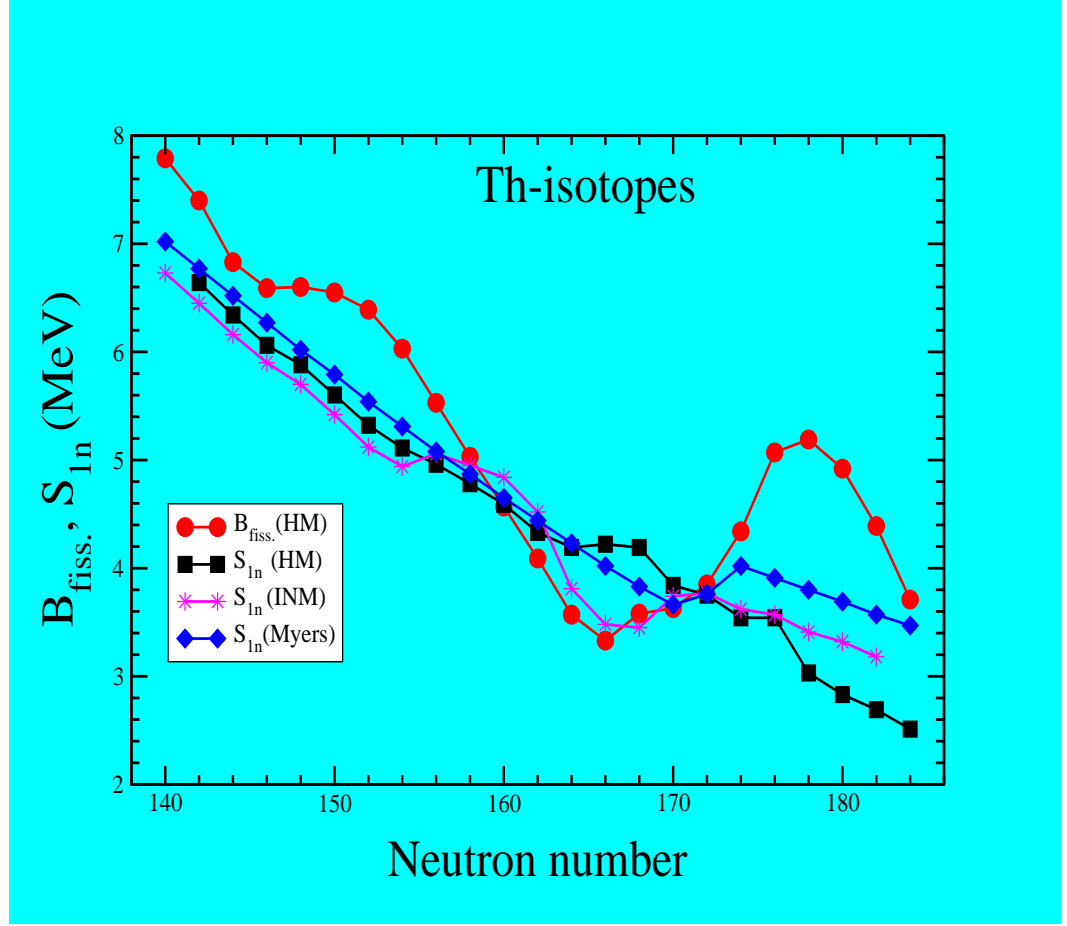


Figure 3: Fission barrier B_f , and binding energy of the last neutron S_{1n} as a function of mass number A for Th isotopes. The B_f are taken from Howard and Möller [25] and S_{1n} are taken from Refs. [25], [24] and [32] for HM, INM and droplet model, respectively. The neutron separation energy predicted by all the three models predict a general pattern. In all the three cases the fission barrier lies above the single particle energy, except for the region $N = 162 - 170$, which will be vulnerable to thermal neutron fission.

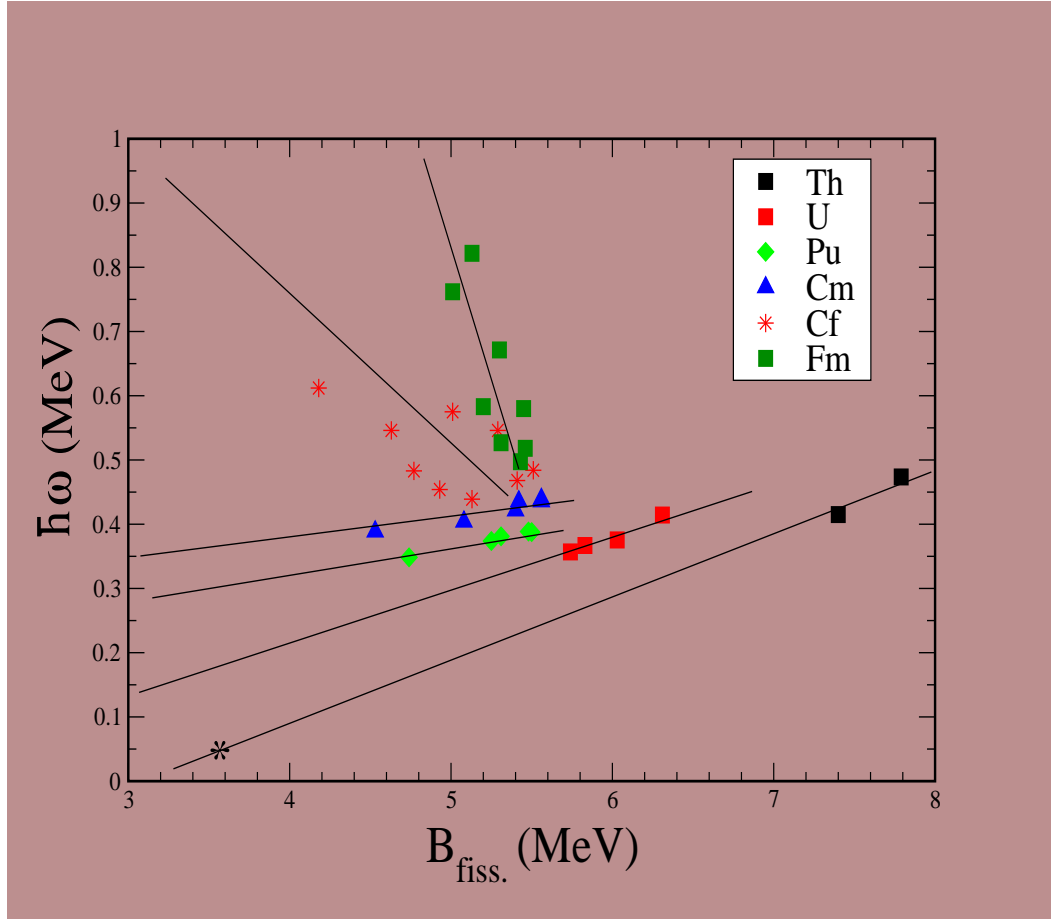


Figure 4: The barrier width $\hbar\omega$ as function of fission barrier B_f . The B_f values and the experimental half-lives taken from Howard and Möller [25] and Ref. [29], respectively have been used to extract the barrier widths of all the known nuclei showing fission decay in the actinide region. This Figure shows that for each element $\hbar\omega$ and B_f show a linear relationship through a straight line behavior depicted in the figure. The star on the line for Thorium in figure, denotes the value of $\hbar\omega$ to be 0.05 MeV for its $B_f = 3.57$ MeV. This linear relationship has been used to obtain $\hbar\omega$ for unknown isotopes by extrapolation shown by straight line.

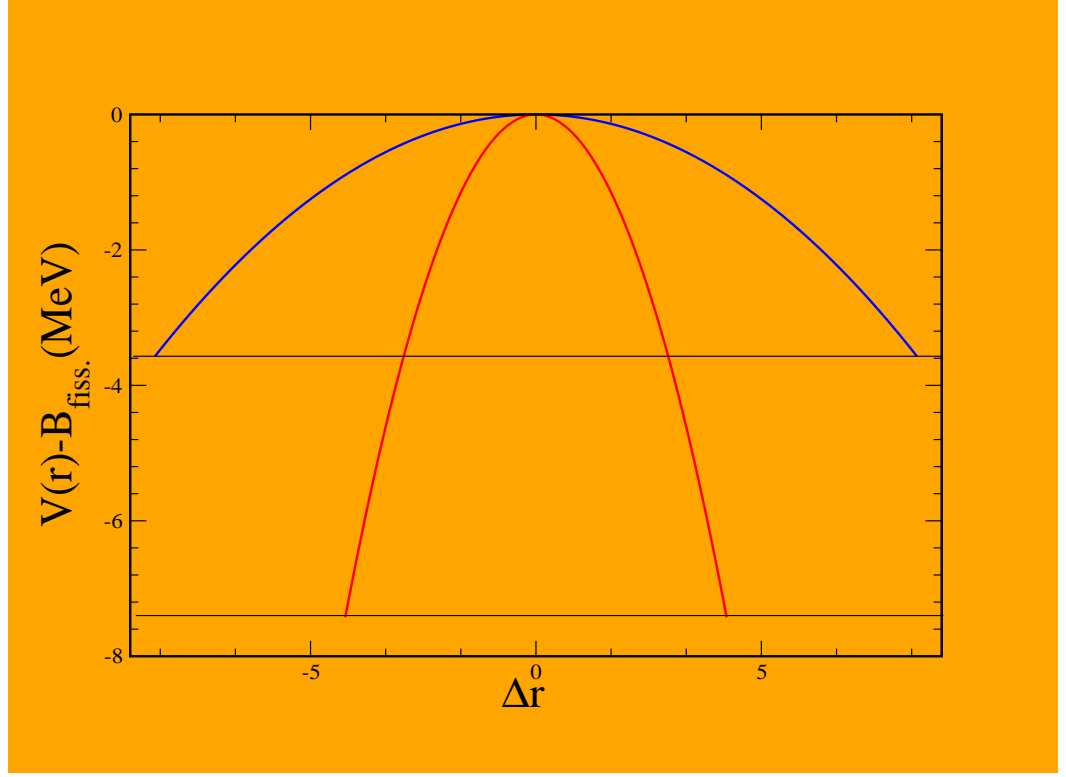


Figure 5: The schematic curve is constructed using fission barrier height B_f and width $\hbar\omega$ to represent the shape of the parabolic fission barrier using the expression $y = -ax^2$. Here $a = \hbar\omega$, $y = d^2V/dr^2$ and $x = \Delta r$ the base width of the parabola, where V is the potential energy. This figure shows that smaller the value of $\hbar\omega$ larger is the width. The normal nuclei like $^{230,232}\text{Th}$ or $^{235,238}\text{U}$ have $\hbar\omega \approx 0.4 - 0.5\text{MeV}$. In the figure, the red curve represents the fission barrier of ^{232}Th and the blue one of ^{254}Th . For the neutron rich superheavy isotope of ^{254}Th , the width is flattened unexpectedly and the height decreases considerably as shown in the blue curve. This flattening of fission barrier makes the nucleus stable against spontaneous fission decay, because of decreasing penetrability. On the other hand, due to the decrease of fission barrier, with a minute inducement by a thermal neutron, fission decay will occur.

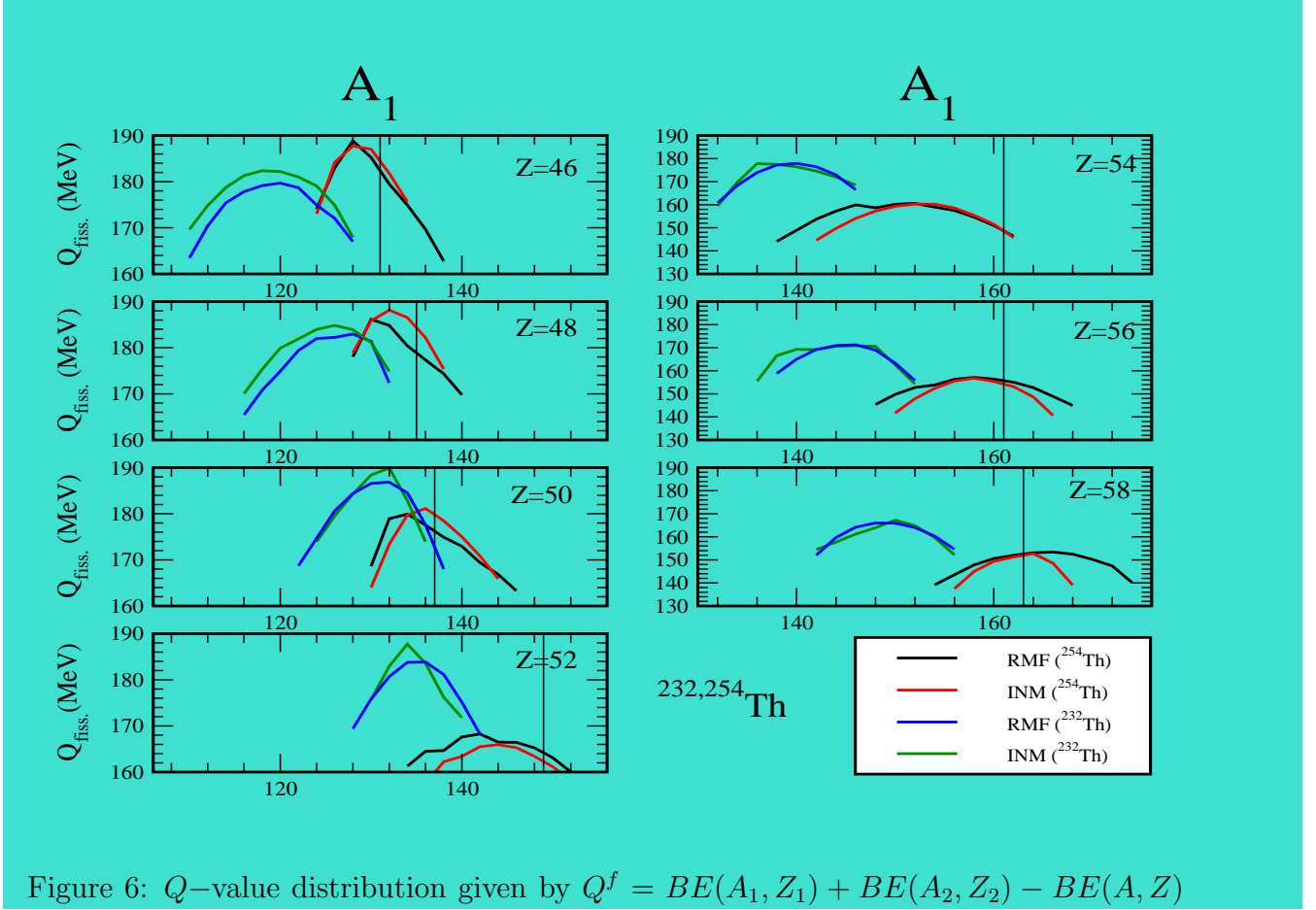


Figure 6: Q -value distribution given by $Q^f = BE(A_1, Z_1) + BE(A_2, Z_2) - BE(A, Z)$ for ^{232}Th and ^{254}Th as a function of A_1 fragment in the binary decay $A \rightarrow A_1 + A_2$. The binding energy used for calculation of Q -value is taken from Ref. [24] and [26] for INM and RMF respectively. The fission yield decreases drastically with increase or decrease of mass number of given element (A_1, Z_1). Therefore, we have shown the distribution in the range 90 to 100% of the peak value in each case. The vertical line marks the neutron drip-line for the corresponding element in each panel. From the figure, it is clear that for ^{232}Th no fragment is produced with isotopes beyond the drip-line. On the other hand, for ^{254}Th a larger number of fragments are predicted to lie away from the drip-line, giving rise to some neutron emission simultaneously, which may be termed as multi-fragmentation fission.